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論文題目	POTENTIALLY TOXIC ELEMENT CONTAMINATION IN MALAYSIAN CLOSED URBAN LANDFILLS: GEOCHEMICAL INDICES AND GEOSTATISTICAL ANALYSIS （マレーシアの閉鎖都市埋立処分場における有害元素汚染の可能性：地球化学的指標と地球統計学的解析）		

（論文内容の要旨）

This thesis comprises of two main parts of analysis, i.e., geochemical indices assessment and geostatistical analysis for the evaluation of potentially toxic elements (PTEs) contamination in closed urban landfills. The thesis is divided into eight chapters with each chapter including fundamental section as follows:

Chapter 1 Introduction

This chapter in brief explained the general and arbitrary background of Potentially Toxic Elements (PTEs) pollution in soil environment, landfills management and environmental risk. Benefits of this research to various sectors, novelty and impact of this work were also covered in detail.

Chapter 2 Literature review

The literature review started with the general issues of PTEs pollution and its relation to human health. It was then narrowed down to PTEs pollution in soil and plant bioaccumulation focusing on PTEs route uptake. Apart from that, waste management schemes on municipal solid waste in Malaysia and Japan were also discussed particularly on the system practiced and technology applied. The literature in relation to the main parts of analysis in this study, i.e., geochemistry (covering PTEs sources, metal stable isotope signatures, indices and mapping technology) and geostatistics (covering background and general workflow) were collected distinctively and presented in this chapter.

Chapter 3 Research site

The closed landfills (sanitary and non-sanitary) were introduced with location map and soil's physico-chemical characteristics of selected closed urban landfills for this research, i.e., Air Hitam (AH) Landfill (closed sanitary) and Ampar Tenang (AT) Landfill (closed non-sanitary) were also presented.

Chapter 4 Method of measurement

This chapter described several sets of methodologies applied for this research. The 10-method design was employed for estimation of ordinary kriging whereas 5 m × 5 m grid design for estimation of advanced kriging. In laboratory analysis part, two segments were exhibited, i.e., sample preparation and instrumental analysis (ICP-MS, pXRF and XRF) whereas in geochemical indices part concisely listed the indices, i.e., C/p index, Er^i-RI, and EF adopted for evaluation of PTEs contamination in soil, water and plant respectively. The last part in this chapter, geostatistical analysis comprehensively described in detail about geostatistical procedures and explanation of two categories, i.e. ordinary kriging and advanced kriging which were conducted by the support of VBA application in Microsoft® Excel.

Chapter 5 Geochemical indices assessment

The geochemical indices assessment results showed that very slight contamination of the soils at both landfills (finding from the analysis of C/p index), with low ecological risk for the stagnant water on both landfills' surface soils (finding from the analysis of Er^i-RI) and most of the PTEs in the plant species at AH landfill were formerly from contaminated surface soil of the closed landfill. A metal-sensitive plant, that is, *Ageratum conyzoides*, was identified as a potential micropollutant

indicator for PTEs contamination in both of the studied closed landfills. This is noticeable when the overall results of PTEs concentration of this study demonstrated that the Cd was not detected in the soil of both landfills, but the concentration was present in the foliar organ of *A. conyzoides* and not in other plant species at both study sites. The highest uptakes in plant for Cd was in foliar organ of *A. conyzoides* (0.934 ppm) which has grown on AT landfill.

Chapter 6 Geostatistics: ordinary kriging

The ordinary kriging analysis in AT landfill showed that Mn, Fe and Pb kriging estimates were classified more accurately compared to Cr, Cu and Cd kriging estimates based on N/S ratio < 0.3 classification. However, AH landfill showed that Cr, Cd and Pb kriging estimates were classified more accurately compared to Mn, Fe and Cu kriging estimates. Using ordinary kriging, we found that the average value of Cr at AH landfill was expected to be over criteria at about 25%, but its ninety-five percentile value was expected to be over criteria at about 75%. It showed that the risk of the element of Cr could not be negligible, although, its risk might be overlooked if average value was utilised in the assessment. Based on ordinary kriging estimates all of the PTEs tested at both sites were below the permissible limits. Moreover, the interpolation area estimated gives useful understanding for monitoring the PTEs contamination by analysing the changes of the PTEs content at both closed landfills.

Chapter 7 Geostatistics: advanced kriging

The advanced kriging analysis, i.e., regression kriging and guess-field kriging efficiency were tested by applying the regression relationship of auxiliary variable to generate estimation data for deriving target variable's spatial structure and kriging estimates. In this study, Pb and Cr data from the analysis of ICP-MS were used for all of the analysis of the advanced kriging. In regression kriging analysis, regression error (regression residual) must be independent from each other and has spatially no correlation. It was shown in the analysis that distribution of regression error possessed spatial correlation. Therefore, application of regression kriging to target variable, i.e., Pb (ICP-MS) using the data of auxiliary variable, i.e., Cr (ICP-MS) was thought not to be appropriate. Guess-field kriging is applicable to the case above. It showed that it was efficient at area where sufficient number of auxiliary variables existed. However, it also showed that guess-field kriging might produce larger uncertainty than ordinary kriging at area where auxiliary variables did not exist. In the case that normal distribution data to be interpolated showed existence of censored data where its normal plot bended at point of non-detection (n.d.) limit of the instrumental method, the application of indicator kriging was suitable as an advanced Kriging. The efficiency of indicator kriging was analysed in occasion that the data contained many n.d. values and the polluted area to be removed should be decided in case of soil remediation.

Chapter 8 Conclusions and recommendations

This chapter concluded the findings of the two main parts of analysis, i.e., geochemical indices assessment and geostatistical analysis as well as recommendations for future work.

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（論文審査の結果の要旨）

本論文はマレーシア都市部における閉鎖埋立処分場の有害元素による汚染可能性を評価するための、地球化学的指標と地球統計学的解析方法の有効性について検討したものである。本論文の学術的意義は、以下の通りである。

1）マレーシアの都市近郊の Air Hitam (AH)埋立処分場と Ampar Tenang (AT)埋立処分場で、土壌や地表水、そこに生えている植物中各種重金属濃度を測定した。その結果、この地域に多い植物であるカッコウアザミの葉中濃度が重金属汚染、特に Cd に敏感であり、土壌の重金属汚染の指標植物として有効であることを示した。また、AH 埋立処分場の植物中重金属濃度は主に表層土壌に由来するが AT 埋立処分場に生える植物の葉中重金属は車の排気ガス中重金属の沈積に由来すると考えられることを明らかにした。

2）Ordinary Kriging(OK)を AH 埋立処分場の土壌中 Cr 濃度に適用した場合、中央値では 25%の面積で規準値を超える可能性があったが、95 パーセンタイル値では 3 倍の 75%の面積で規準値を超える可能性が示された。このことは土壌中 Cr 濃度の平均値や中央値を使って Cr のリスクを評価した場合、大きなリスクを見逃してしまう可能性があること、OK を用いると、平均値や中央値では現れてこないリスクを見いだすことも可能であることを示した。

3）土壌汚染濃度の推定精度を上げるために、推定量以外の変数を補助変数として用いる高度 Kriging 法として Regression Kriging (RK) と Guess-Field Kriging (GFK) の利用を試みた。Cr と Pb の土壌中濃度の全量分析値の間には相関係数 0.73 の相関が観測されたので、この相関関係を用いて、Pb の未観測領域の値を、まず、Cr の観測値を用いた RK で推定したが、推定残差に空間的相関があり、RK の適用は不適当であることを示した。そのため GFK を適用したが、Cr の観測データが存在する地域周辺では OK に比べ Pb 濃度の平均推定誤差が減少したが、Cr データが存在しない推定領域周辺では GFK の方が平均推定誤差が増大する場合があることを示した。また、検出限界以下のデータが多数存在して OK が適用できないような場合には、Indicator Kriging の適用が効果的であることを示した。

以上のように、本論文は、閉鎖埋立処分場における土壌重金属汚染のリスク評価の精度を上げるために、植物中濃度を利用する方法や高度 Kriging 法を適用することの有効性を検証したものであり、土壌中重金属濃度などの十分な観測データを得ることが困難なマレーシア等の開発途上国においては、実用性の高い研究であり、また、開発途上国における土壌汚染リスク管理手法の確立に大きく貢献するものであって、学術上、実際上寄与するところが大きい。よって、本論文は博士（工学）の学位論文として価値あるものと認める。また、令和２年２月１０日、論文内容とそれに関連した事項について試問を行った結果、合格と認めた。なお、本論文は、京都大学学位規程第 14 条第 2 項に該当するものと判断し、公表に際しては、（令和３年３月３１日までの間）当該論文の全文に代えてその内容を要約したものとすることを認める。